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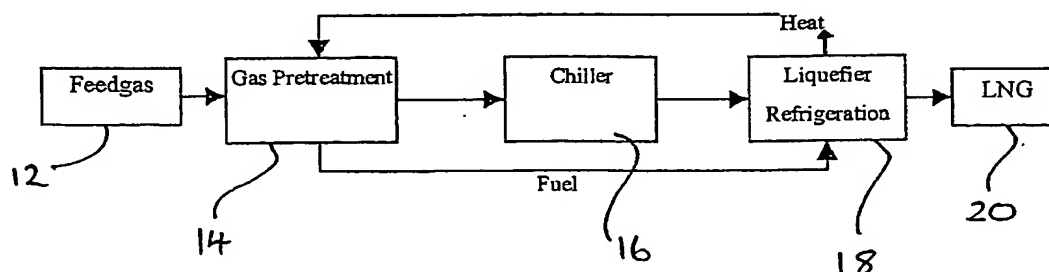
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(54) Title: A REFRIGERATION PROCESS AND THE PRODUCTION OF LIQUEFIED NATURAL GAS



(57) Abstract: Process and apparatus for the production of liquefied natural gas utilising a refrigeration cycle, characterised by the steps of: i) Pre-treatment of a natural gas stream; ii) Chilling of either or both of the resulting pre-treated gas stream or a refrigerant gas stream within the refrigeration cycle; and iii) Liquefaction of the natural gas.

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## "A Refrigeration Process and the Production of Liquefied Natural Gas"

### Field of the Invention

The present invention relates to a refrigeration process. More particularly, the refrigeration process of the present invention has particular application in the  
5 production of liquefied natural gas.

### Background Art

Traditional processes for the production of liquefied natural gas (hereinafter "LNG") comprise, in broad terms, a natural gas pre-treatment stage and a gas liquefaction stage. The pre-treatment stage is required to remove components of  
10 the gas stream that will freeze solid at cryogenic temperatures. Examples of components removed for this reason are carbon dioxide, hydrogen sulphide, heavy hydrocarbons and water. Carbon dioxide and/or hydrogen sulphide is typically removed in an absorption process (for example using amine) and/or membrane process; heavy hydrocarbons removed by cooling and condensing,  
15 and water removed in a dehydration process (for example using molecular sieves). Such pre-treatment may either require or cause the gas to be heated to about 50°C.

The liquefaction stage of the process comprises both cryogenic heat exchange and refrigeration. The pre-treatment stage provides 'sweet dry' gas which is  
20 passed through a heat exchanger and expansion valve, where it is cooled to about -150°C (depending upon gas composition and storage pressure), liquefied and transferred to storage. A variety of refrigeration methods using various refrigerants and processes are known.

In one example of the prior art (typically for small scale plants) the refrigeration  
25 step comprises each of a standard compression, cooling by air or water and an expander cycle, in which most refrigeration is provided by the isentropic expansion of a recycle stream. A turbo expander-compressor is used to recover power from gas expansion and the refrigerant is further compressed in main gas

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driven booster compressors. Warm refrigerant is pre-cooled by cold refrigerant gas prior to entering the expander cycle so that the necessary cryogenic temperatures can be achieved.

5 In another example of the prior art (typically for larger plants), two refrigerant cycles are provided. Each cycle has its own compressor drive (traditionally using gas turbines but could equally use electric drives powered by gas turbine generators). The "first" cycle is used to pre-cool the natural gas as well as pre-cool the "second" lower temperature cycle. Refrigerant for the first cycle typically uses propane or mixed refrigerant.

10 Typically employed processes for the production of LNG as described above presently have substantial energy requirements for cooling and liquefaction of the natural gas. Alternatively, if a more energy efficient process is selected, that process will be very expensive in terms of initial capital costs. This energy is supplied by mechanical drives that use prime movers, such as gas turbines, gas  
15 engines and/or electric motors, to drive compressors for the necessary refrigeration processes. The prime movers are inherently very inefficient and are known to typically convert only 25 – 40% of the energy supplied as fuel into useful compressive work for the refrigeration process. The majority of energy is lost to atmosphere in the form of heat. As such, presently available processes for LNG  
20 production are very inefficient.

In known LNG processes the feed natural gas is typically pre-treated to remove carbon dioxide, heavy hydrocarbons and water prior to liquefaction. This pre-treatment requires heating in a solvent absorption or membrane system. As a result, further cooling energy is then required to liquefy the natural gas.

25 The process for the production of liquefied natural gas of the present invention has as one object thereof to overcome substantially the abovementioned problems of the prior art, or to at least provide a useful alternative thereto.

Throughout the specification, unless the context requires otherwise, the word "comprise" or variations such as "comprises" or "comprising", will be understood to

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imply the inclusion of a stated integer or group of integers but not the exclusion of any other integer or group of integers.

The preceding discussion of the background art is intended to facilitate an understanding of the present invention only. It should be appreciated that the discussion is not an acknowledgement or admission that any of the material referred to was part of the common general knowledge in Australia or any other country and/or region as at the priority date of the application.

### Disclosure of the Invention

In accordance with the present invention there is provided a process for the production of liquefied natural gas utilising a refrigeration cycle, the process characterised by the steps of:

- i) Pre-treatment of a natural gas stream;
- ii) Chilling of either or both of the resulting pre-treated gas stream or a refrigerant gas stream within the refrigeration cycle; and
- 15 iii) Liquefaction of the natural gas.

Preferably, the chilling step is driven at least in part by waste heat from the liquefaction step. The waste heat may comprise hot jacket water and/or hot exhaust gases from the main gas engine or turbine driven compressor. Additionally, heat may also be provided from one or more of the group of prime movers, compressors, burning of flare or other waste gases or liquids, and solar power.

Still preferably, waste heat from the liquefaction step is utilised, at least in part, in the gas pre-treatment step.

The chilling step may further condense certain components of the pre-treated natural gas stream. Components of the natural gas stream condensed in this manner may include water, heavy hydrocarbons and/or carbon dioxide.

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Further preferably, the chilling step cools the gas stream to a temperature of between about  $-80^{\circ}\text{C}$  and  $10^{\circ}\text{C}$ . The chilling of the pre-treated gas stream is preferably conducted in a number of stages so as to allow the selective condensation and removal of various components thereof.

- 5 The chilling of the refrigerant gas stream may cause some components in the refrigerant gas to condense. The liquid thus formed may be pumped and flashed to improve efficiency as in a conventional mixed refrigerant cycle.

Still further preferably, the chilling step utilises either a lithium bromide or an ammonia absorption chiller.

- 10 In one form of the invention either a turbo-expander or 'JT' valve or nozzle device is added between the chilling step and the liquefaction step to further cool the natural gas stream.

- In accordance with the present invention there is further provided apparatus for the production of liquefied natural gas, the apparatus comprising an absorption  
15 and/or membrane package for carbon dioxide removal, a dehydration package for water removal, a liquefaction package, at least one chiller and at least one refrigerant compressor package, the chiller being arranged so as to chill the natural gas stream to be liquefied.

- In one form of the invention the liquefaction package further comprises the chiller  
20 arranged to chill a pre-treated natural gas stream from the solvent absorption and dehydration packages prior to passing that gas stream to a cryogenic heat exchanger.

- In another form of the present invention the chiller is located before, or as a part of, the amine and/or membrane packages so as to assist in pre-treatment of the  
25 natural gas stream. The chiller may comprise one or more chiller stages.

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In a yet further form of the invention the chiller is located in the refrigeration cycle to improve the efficiency thereof. The chiller may be located in both the natural gas stream and refrigeration package, or in either one thereof.

Preferably, the chiller is driven by waste heat from the or each refrigerant compressor packages. This waste heat may also be directed to the amine package for amine regeneration and/or to the dehydration package for regeneration of molecular sieves used therein.

The chiller may be provided in the form of either an ammonia or lithium bromide absorption chiller. The ammonia absorption chiller preferably cools the gas stream to about -30 to -80°C whereas the lithium bromide absorption chiller cools the gas stream to about 0 to 10°C.

A turbo-expander or "JT" valve or nozzle device may be added downstream of the chiller.

In accordance with the present invention there is still further provided a refrigeration process in which waste heat is utilised to chill a process stream thereby reducing the refrigeration load.

In one form of the present invention the refrigeration process is utilised in an air separation plant. In a further form of the invention the refrigeration process is employed in an LPG extraction process. In a still further form of the present invention, the refrigeration process is employed to pre-treat the gas.

### **Brief Description of the Drawings**

The present invention will now be described, by way of example only, with reference to one embodiment thereof and the accompanying drawings, in which:-

Figure 1 is a schematic flow chart of a process for the production of liquefied natural gas in accordance with the present invention;

Figure 2 is a schematic representation of one embodiment of the process

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of Figure 1;

Figure 3 is a pressure enthalpy diagram for the process of the present invention using an ammonia absorption chiller in which the chilling step cools a natural gas stream to about -50°C;

5      Figure 4 is a graph of Temperature vs Enthalpy in the process of Figures 2 and 3, demonstrating the effect of the absorption chiller on overall cooling load; and

10      Figure 5 is a schematic flow chart of a process for the production of liquefied natural gas in accordance with a second embodiment of the present invention.

### **Best Mode(s) for Carrying Out the Invention**

In Figure 1 there is shown a process 10 for the production of liquefied natural gas in accordance with the present invention. The process 10 broadly comprises passing a natural gas feed gas 12 to a gas pre-treatment step 14, after which the  
15      gas stream is passed to a chiller 16. The chiller 16 cools the gas stream to at about -50°C prior to the gas stream passing to a liquefaction stage 18, finally producing a liquefied natural gas ("LNG") product 20.

As shown in Figure 1; waste heat from the liquefaction stage 18 is utilised by both the chiller 16 and the pre-treatment step 14.

20      In Figure 2 there is shown the process 10 in greater detail than that of Figure 1.

The natural gas stream 12 is subjected to a pre-treatment step 14 comprising an amine package 22 and a dehydration package 24. The amine package 22 and the dehydration package 24 remove carbon dioxide and water from the natural gas stream 12 respectively. Broadly speaking, the pre-treatment step 14 is  
25      required to remove components in the natural gas stream 12 that would otherwise freeze at cryogenic temperatures experienced in the liquefaction step 18. The pre-treatment step 14 normally requires the natural gas stream 12 to be heated to

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about 50°C. As such, this step demands more cooling and more energy to ultimately reach liquefaction temperature in the subsequent liquefaction step 18.

The liquefaction step 18 comprises at least the majority of a liquefaction package 26 shown in Figure 2, the liquefaction package 26 comprising a main cryogenic heat exchanger 28 and one or more expander compressors 30 together with a refrigeration cycle 32. The refrigeration cycle 32 further comprises one or more refrigerant compressor packages 34.

The liquefaction package 18 provides LNG that is passed to one or more LNG tanks 36 via an LNG separator 39.

10 The sweet dry natural gas produced by the pre-treatment step 14 passes through the heat exchanger 28 and an expansion valve 38, where it is cooled to around -150°C and liquefied prior to passing to the LNG tanks 36. The LNG separator produces a small volume of flash gas 39 that is used as make-up gas for the refrigeration cycle 32, as a regeneration gas 40 and finally as a fuel gas 41 for the compressor drives 34.

The refrigeration cycle 32 comprises a multi-stage compression, air or water cooling and expander cycle, with most refrigeration produced by isentropic expansion of a recycle stream. Power from gas expansion is recovered in a turbo expander-compressor and the refrigerant is further compressed in the main gas engine or turbine driven booster compressors. Warm refrigerant is precooled by cold refrigerant gas prior to entering the expander so that the required cryogenic temperature in the heat exchanger 28 can be achieved.

The chiller 16 is provided in-line between, or upstream of, the pre-treatment step 14 and the liquefaction package 18. The chilling step 16 may be achieved by either of a lithium bromide absorption chiller, cooling the natural gas stream to about 10°C, or an ammonia absorption chiller, cooling the natural gas to about -50°C, or may be a combination of these methods. This chilling of the natural gas stream prior to the heat exchanger 28 reduces significantly the load on the



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liquefier/refrigeration plant by, in the experience of the applicants, as much as 50% compared with the prior art.

The chiller step 16 utilises waste heat 42, comprising hot jacket water and/or hot exhaust gases, from the main gas engine compressor drives 34. This heating  
5 system may also be used to regenerate the amine and/or preheat the natural gas stream prior to entering membranes and/or heat regeneration gas required for the molecular sieves of the dehydration package 24. Hot dry refrigerant gas from the compressor discharge may also be used to regenerate the molecular sieves of the dehydration package 24, prior to that same gas being used as fuel for the  
10 compressor drives 34.

Additional heat may be utilised in the chiller step 16, such as may be available as waste heat from other prime movers for example those used for power generation, heat from compression from the burning of flare or other waste gases or liquids, solar power and the like.

15 It is also to be understood that, dependent upon the composition of the natural gas stream 12, another benefit of the process 10 of the present invention is that the chilling step 16 may condense some components, including heavy hydrocarbons, LPG's, water, hydrogen sulphide and/or carbon dioxide. These condensed components can either be a useful product stream or may assist in the  
20 pre-treatment process itself. Additionally, the flash gas 39 from the LNG separator 37 is high in nitrogen, thereby improving the heating value of the LNG product 20. Further, the flash gas 39 is bone dry making it especially suitable for regeneration gas 40 and making it especially suitable as fuel gas 41 in the compressor drives 34 due to its high methane number.

25 In Figure 3 there is shown a pressure enthalpy diagram for the process 10 of the present invention utilising an ammonia absorption chiller cooling the natural gas stream to about -50°C, followed by an expander or "JT" valve 38, as shown in Figure 2, to further pre-cool the natural gas stream. It is envisaged that a compressor, for example a vacuum compressor (not shown), may also be added  
30 to the ammonia circuit to further pre-cool the natural gas.

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In Figure 4 there is shown a graph of temperature vs enthalpy from the heat exchanger 28 demonstrating the significant reduction in cooling load on the heat exchanger as a result of the presence of the absorption chiller 16 which has cooled the natural gas stream to about  $-50^{\circ}\text{C}$ .

- 5 It is envisaged that more than a single chiller step 16 may be utilised. The or each chiller step 16 may additionally be driven by sources of heat other than the refrigerant compressor packages described hereinabove.

It is further envisaged that the or each chiller step 16 may utilise fluids other than the ammonia and lithium bromide described hereinabove.

- 10 In Figure 5 there is shown a process 100 for the production of LNG in accordance with a second embodiment of the present invention. The process 100 is substantially similar to the process 10 described hereinabove and like numerals denote like parts and steps.

- 15 Importantly, a number of chillers 102 are provided in the process stream, each being driven by waste heat from the refrigeration cycle 32. The chillers 102 are placed within the gas pre-treatment step 14 directly after each of carbon dioxide removal and drying, and immediately prior to the heat exchanger 28 of the refrigeration cycle 32. As noted previously, this staged chilling of the natural gas stream 12 allows selective condensation and removal of various components thereof. Within the refrigeration cycle 32 a chiller 102 is used to chill mixed refrigerant.

- 25 The processes 10 and 100 for the production of LNG each utilise waste heat from the refrigeration cycle to generate heat or cold as required, thereby increasing the efficiency of the LNG production process when compared with prior art processes. For example, prior art LNG processes lose energy by way of waste heat to atmosphere. The present invention utilises waste heat to chill the natural gas and/or refrigerant, thereby improving the efficiency of the process, reducing capital and operating costs, reducing greenhouse gas emissions and simplifying

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the process. Alternatively, a similar efficiency to that of the prior art processes may be achieved at a lower capital cost.

It is envisaged that the process of the present invention may be applied broadly to refrigeration processes, including those used in air separation plants and LPG  
5 extraction processes, thereby providing similar benefits with regard to utilisation of waste heat. Each of these processes require refrigeration and waste heat can again be utilised to chill the stream, thereby improving efficiency and reducing costs.

It is further envisaged that the refrigeration process described above may be used  
10 to refurbish existing inefficient LNG or air separation plants.

Modifications and variations such as would be apparent to the skilled addressee are considered for within the scope of the present invention.

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## Claims

1. A process for the production of liquefied natural gas utilising a refrigeration cycle, the process characterised by the steps of:
  - i) Pre-treatment of a natural gas stream;
  - 5 ii) Chilling of either or both of the resulting pre-treated gas stream or a refrigerant gas stream within the refrigeration cycle; and
  - iii) Liquefaction of the natural gas.
2. A process according to claim 1, wherein the chilling step is driven at least in part by waste heat from the liquefaction step.
- 10 3. A process according to claim 2, wherein the waste heat comprises hot jacket water and/or hot exhaust gases from the main gas engine or turbine driven compressor.
4. A process according to claim 1 or 2, wherein heat is provided from one or more of the group of prime movers, compressors, burning of flare or other  
15 waste gases or liquids, and solar power.
5. A process according to any one of the preceding claims, wherein waste heat from the liquefaction step is utilised, at least in part, in the gas pre-treatment step.
6. A process according to any one of the preceding claims the chilling step  
20 condenses certain components of the pre-treated natural gas stream.
7. A process according to claim 6, wherein components of the natural gas stream condensed in this manner include one or more of water, heavy hydrocarbons and/or carbon dioxide.

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8. A process according to any one of the preceding claims, wherein the chilling step cools the gas stream to a temperature of between about  $-80^{\circ}\text{C}$  and  $10^{\circ}\text{C}$ .
9. A process according to any one of the preceding claims, wherein the chilling of the pre-treated gas stream is conducted in a number of stages so as to allow the selective condensation and removal of various components thereof.
10. A process according to any one of the preceding claims, wherein the chilling of the refrigerant gas stream causes some components in the refrigerant gas to condense, the liquid thus formed being pumped and flashed to improve efficiency as in a conventional mixed refrigerant cycle.
11. A process according to any one of the preceding claims, wherein the chilling step utilises either a lithium bromide or an ammonia absorption chiller.
12. A process according to any one of the preceding claims, either a turbo-expander or 'JT' valve or nozzle device is added between the chilling step and the liquefaction step to further cool the natural gas stream.
13. An apparatus for the production of liquefied natural gas, the apparatus characterised by an absorption and/or membrane package for carbon dioxide removal, a dehydration package for water removal, a liquefaction package, at least one chiller and at least one refrigerant compressor package, the chiller being arranged so as to chill the natural gas stream to be liquefied.
14. An apparatus according to claim 13, wherein the liquefaction package further comprises the chiller arranged to chill a pre-treated natural gas stream from the solvent absorption and dehydration packages prior to passing that gas stream to a cryogenic heat exchanger.
15. An apparatus according to claim 13 or 14, wherein the chiller is located before, or as a part of, the amine and/or membrane packages so as to assist in pre-treatment of the natural gas stream.

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16. An apparatus according to any one of claims 13 to 15, wherein the chiller comprises one or more chiller stages.
17. An apparatus according to any one of claims 13 to 16 wherein the chiller is located in the refrigeration cycle to improve the efficiency thereof.
- 5 18. An apparatus according to any one of claims 13 to 16, wherein the chiller is located in both the natural gas stream and refrigeration cycle, or in either one thereof.
19. An apparatus according to any one of claims 13 to 18, wherein the chiller is driven by waste heat from the or each refrigerant compressor packages.
- 10 20. An apparatus according to claim 19, wherein waste heat is also directed to the amine package for amine regeneration and/or to the dehydration package for regeneration of molecular sieves used therein.
21. An apparatus according to any one of claims 13 to 20, wherein the chiller is provided in the form of either an ammonia or lithium bromide absorption chiller.
- 15 22. An apparatus according to claim 21, wherein the ammonia absorption chiller cools the gas stream to about  $-30$  to  $-80^{\circ}\text{C}$  whereas the lithium bromide absorption chiller cools the gas stream to about  $0$  to  $10^{\circ}\text{C}$ .
23. An apparatus according to any one of claims 13 to 22, wherein a turbo-expander or "JT" valve or nozzle device is added downstream of the chiller.
- 20 24. A refrigeration process in which waste heat is utilised to chill a process stream thereby reducing the refrigeration load.
25. A refrigeration process according to claim 24, wherein the refrigeration process is utilised in either of an air separation plant or an LPG extraction process.

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26. A refrigeration process according to claim 24 or 25, wherein the refrigeration process is employed to pre-treat the gas.

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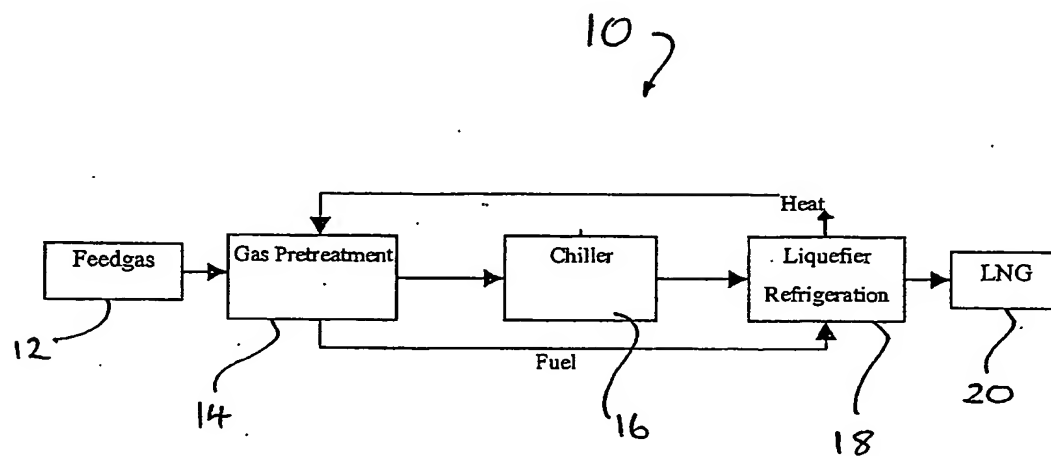


Figure 1



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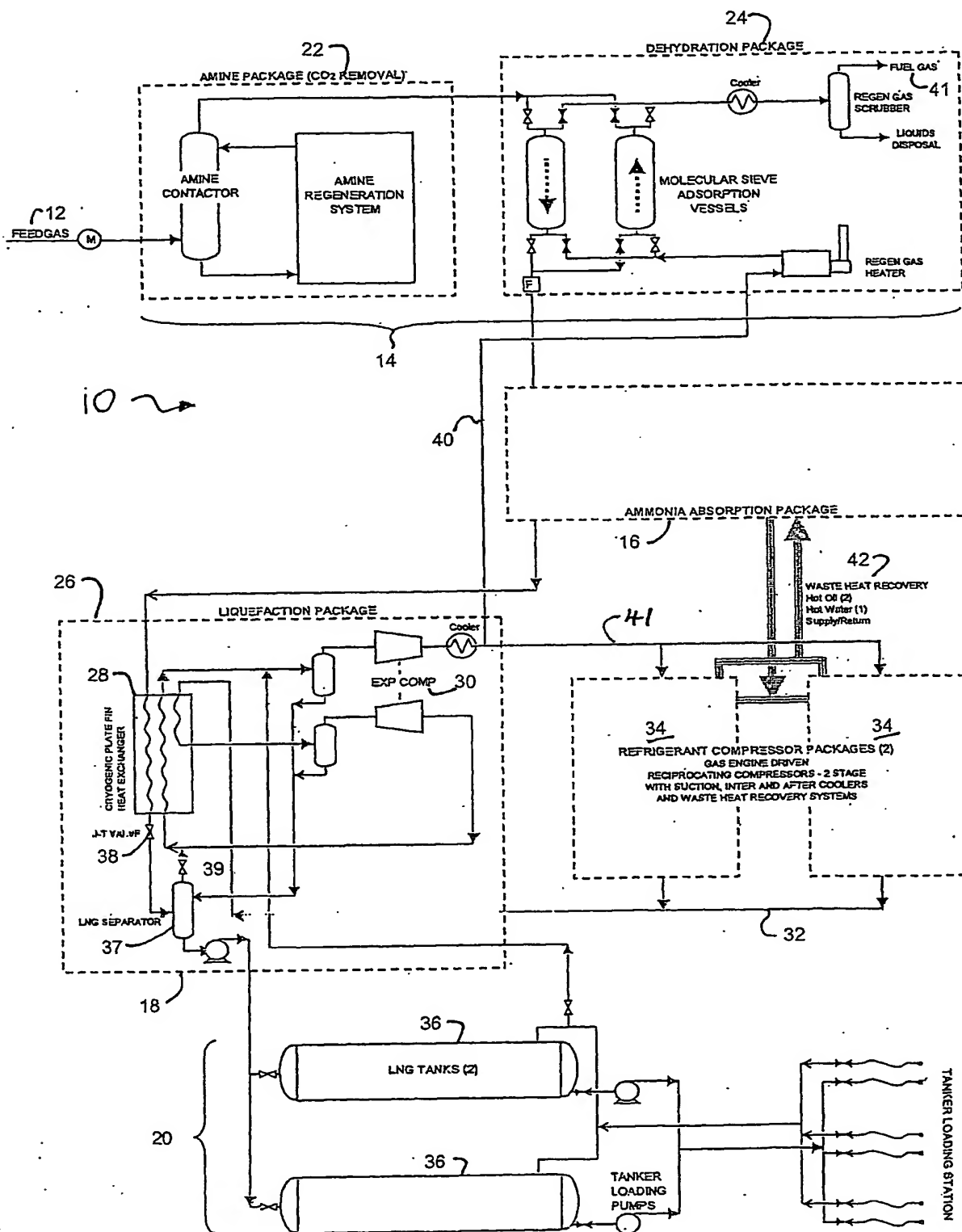


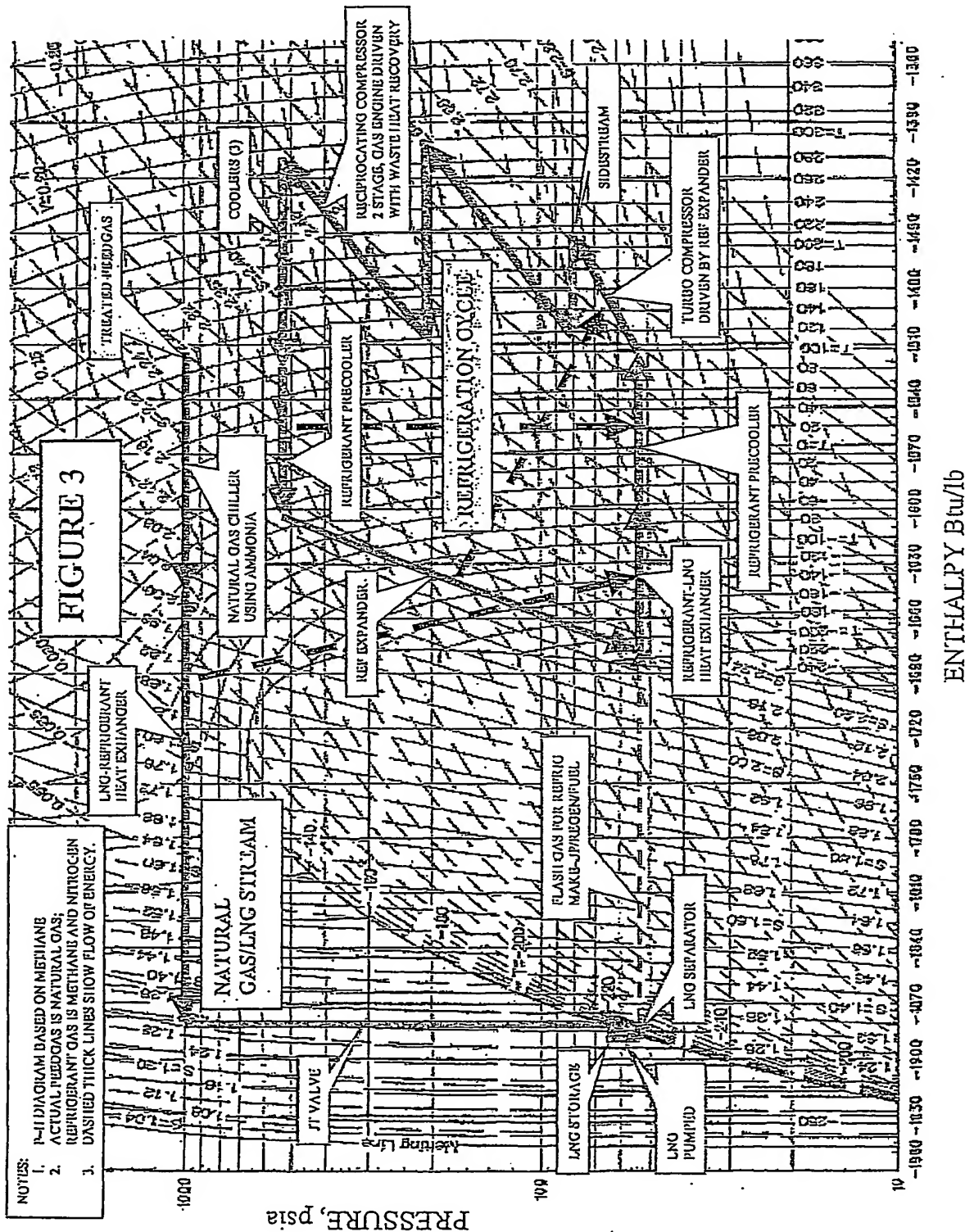
FIGURE 21

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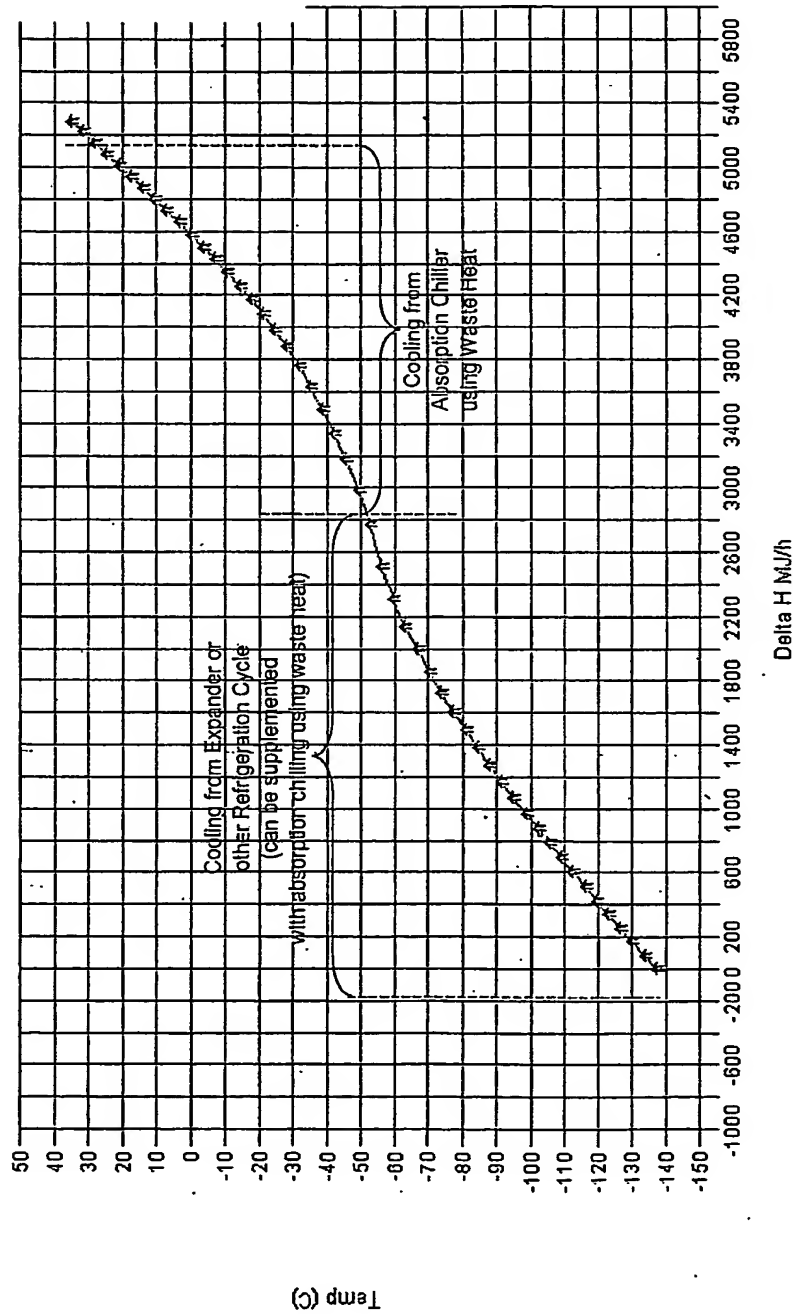
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**FIGURE 4**  
Exchanger 2 Heat Curve



↑ Stream 3

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The diagram illustrates a 12 Feedgas liquefaction process. The feed gas enters from the left, labeled '12 FEEDGAS'. It passes through a 'CO<sub>2</sub> REMOVAL' unit, then an 'AMMONIA CHILLER' (represented by a circle with a wavy line). The gas then enters a vertical vessel where 'WATER HHC' is removed. The gas then passes through a 'DRYER' and another 'AMMONIA CHILLER'. The final product is 'CONDENSATE & LPG', which is collected in a vertical vessel. The gas then enters a 'REGEN/FUEL GAS (COLD RECOVERY)' unit. The gas then passes through a 'JT VALVE' and enters a 'LNG HEAT EXCHANGER'. The gas then enters an 'AMMONIA ABSORPTION SYSTEM'. The gas then enters a 'GAS TURBINE' which produces 'WASTE HEAT'. The gas then enters a 'COMPRESSOR' which is part of a 'MIXED REFRIGERANT SYSTEM'. The gas then passes through two 'COOLERS' and an 'AMMONIA CHILLER'. The gas then enters the 'LNG HEAT EXCHANGER' again. The gas then enters a vertical vessel where 'WATER HHC' is removed. The gas then enters a 'DRYER' and another 'AMMONIA CHILLER'. The final product is 'CONDENSATE & LPG', which is collected in a vertical vessel. The gas then enters a 'REGEN/FUEL GAS (COLD RECOVERY)' unit. The gas then passes through a 'JT VALVE' and enters the 'LNG HEAT EXCHANGER'.

FIGURE 5